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Experience with a General Gamma-Ray Isotopic Analysis Approach

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The gamma-ray data analysis methodology originally developed for the MGA code to determine the relative detection efficiency curve may also be used to determine the relative amounts of the isotopes being measured. This analysis approach is based on the fact that the intensity of any given gamma ray from a sample is determined by the amount of the emitting isotope present in the sample, the emission probability for the gamma ray being measured, the sample self attenuation, the attenuation due to absorbers between the sample and detector, and the detector efficiency. An equation can be written that describes a measured gamma-ray peak intensity in terms of these parameters. By selecting appropriate gamma-ray peaks from the isotopes of interest, we can solve a set of equations for the values of the parameters in any particular measurement including the relative amounts of the selected isotopes. The equations representing the peak intensities are very nonlinear and require an iterative least squares method to solve. We have developed software to ensure that during the iterative process the parameters stay within their appropriate ranges and converge properly in solving the set of equations under various measurement conditions. We have utilized and reported on this approach for determining the plutonium isotopic abundances in samples enriched in Pu-238 and to determine the U-235 enrichment of uranium samples in thick-walled containers. Recently, we have used this approach to determine the plutonium isotopic abundances of plutonium samples in thick-walled containers. We will report on this most recent application, and how this general approach can be adapted quickly to any isotopic analysis problem.

1. INTRODUCTION

The basis for the measurement of the amount of any radioactive isotope by gamma-ray spectrometry is given by

$$I = p \times X \times \varepsilon,$$

where

I = the measured intensity of a gamma ray at a specific energy,

p = the emission probability of the gamma-ray being measured,

X = the disintegration rate of the radioactive isotope in the sample emitting the measured gamma ray, and

ε = the detection or counting efficiency at a specific energy.

The measured peak intensity, and the disintegration rate of the radioactive isotope, X, are related by the values of ε and p.

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The emission probability is a fundamental constant related to the nuclear properties of the isotope being measured. The value of the detection efficiency, ε, depends on a number of factors related to the sample and detector setup.

For uniform solution samples whose volume and counting geometry can be controlled, the absolute counting efficiency for any gamma ray can be determined by calibration. However, many other samples, particularly solid samples, possess characteristics that are unknown or uncontrolled. The counting geometry is not reproducible and the attenuation of the gamma rays by the sample and sample container are unknown. In these situations, an absolute counting efficiency cannot be used, but we can determine the relative counting efficiency from the measured spectrum itself, which allows us to determine the relative amounts of any two isotopes.

To do this, we use a functional form, which was developed for MGA[1] that describes three major components involved in the detection process and the determination of the relative efficiency. These are 1) the detector efficiency, 2) absorption by the steel container walls, and 3) self-attenuation by the materials in the sample. The advantage of basing the functional form on known physical processes is that its shape is determined not only by the data but also by the physical-interaction processes. The variables involved in the three processes are determined from the gamma-ray spectrum itself. The following equation is used to describe these processes as a function of energy:

$$I_j \equiv \sum_{k=1}^{N} (p_{j,k} \times X_k) \times \exp(-\mu_{Abs}(E_j) \times Abs) \times (1 - \exp(-\mu_{Smp}(E_j) \times Smp)) / (\mu_{Smp}(E_j) \times Smp) \times \varepsilon_j^0 \times (1 + bE_j + cE_j^2)$$

Where I_j's are the intensities of selected peaks in the spectrum from the isotopes of interest and X_k are the unknown disintegration rates of these isotopes. The emission probabilities are given by the terms $p_{j,k}$ for each peak j belonging to isotope k. The absorption coefficients, μ_{Abs} and $\mu_{Smp,}$ at a given peak energy are for the attenuation by the sample container and the sample. The container wall thickness, *Abs,* and the sample thickness, *Smp*, are treated as unknown variables. The final term in the equation, ε^0, is the estimated relative detector efficiency for peak j where b and c are unknown variables in a quadratic function used to account for small variations in the efficiency. The term, ε^0, may be calculated using the dimensions of the detector. This equation is very nonlinear in form and therefore the variables must be solved by an iterative least-squares method. Note that in addition to determining the relative detection efficiency for the measurement, the solution to the developed equations provides the relative amounts of the isotopes involved, when corrected for half-lives.

Previously, we have written software in which this methodology was "hardwired" for application to specific isotopic problems, such as the analysis of plutonium samples[2] with enhanced amounts of ^{238}Pu and samples[3] of enriched uranium with thick container walls. In this paper, we report on software developed that allows this methodology to be adapted by the user to most isotopic measurement problems.

2. DESCRIPTION OF THE GENERAL ANALYSIS SOFTWARE

There are two major steps in solving the equation described above: 1) Determination of the gamma-ray peak intensities, and 2) Determination of the relative amounts of the isotopes contributing to the selected gamma rays. Users, if they choose, may provide their own peak

intensities to the second step. We have written a subroutine to fit overlapping x- and gamma-ray peaks in a peak grouping according to specifications given by the user. The user provides the following information in a text file, which can be created and edited with a simple text editor.

a. The beginning energy in keV of the region containing the peak or peaks to be analyzed, and the width in keV of the background window below the region.
b. The ending energy in keV of the region, and the width in keV of the background window above the region.
c. The number of x- and/or gamma-ray peaks in the specified region. If there is a single peak, the background is subtracted from the specified region and the sum of the net counts is made to determine the peak area.
d. For each peak,
   - its energy in keV,
   - its emission probability,
   - its the Lorentzian line width in eV for x rays,
   - an integer value indicating if the position of the peak is to be freed or fixed relative to another peak in the peak grouping during the fitting process,
   - an integer value indicating whether the height of the peak is to be freed or fixed relative to another peak during the fitting process using the given emission probabilities, and
   - an integer value indicating whether the peak is to be used to determine the relative amount of the isotope from which it was emitted.
e. Three integer values that specify whether the full-width-at-half-maximum, the low-energy tail amplitude, and the low-energy tail slope are to be freed or fixed in the fitting process.

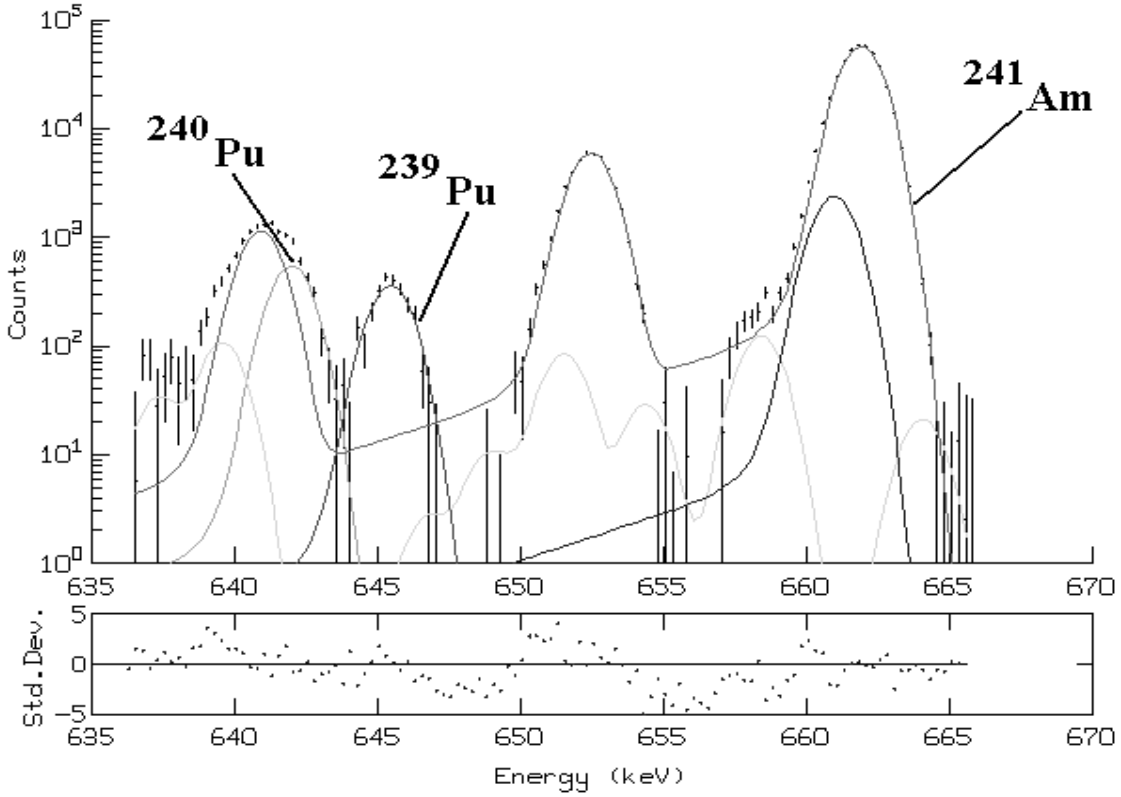

Fig. 1 This plot shows a fit to net plutonium gamma-ray data in the 640-keV region to determine peak areas using prescribed analysis information that relates selected peaks to others in the fit.

The user provides the information described above for each peak or peak groupings used in the analysis. For those peak regions with more than one peak, a peak-fitting routine similar to GRPANL[4] is used that fits a Gaussian peak shape with an exponential low-energy tail to each of the specified peaks. The fitting determines the net peak area for each of the specified peaks along with an error that is based on counting statistics and the quality of the fit. A fit to the 640-keV region in a plutonium gamma-ray spectrum is shown in Figure 1.

For the second part of the analysis to determine the relative disintegration rates for the isotopes of interest, the user selects which peak intensities are to be used for the equations described above. The number of equations must be equal to or greater than the number of unknowns. The unknowns are the relative disintegration rates for the isotopes involved, the sample mass thickness, the absorber mass thickness (usually the container wall), and the slope and curvature of the detector's relative efficiency.

For the peak intensities selected for use in the analysis, the user specifies the isotope that it belongs to as part of the text file described above. In the same text file, the user specifies the half-lives of the isotopes being analyzed for.

To solve the set of equations created from the selected peak intensities, the user also specifies the composition and estimated mass thickness of the sample, and the compositions and mass thickness for up to three absorbers. The thickness of at least two of the three absorbers must be known. Finally, the user specifies the coefficients from a log-log fit to the detector's relative efficiency normalized to 1.0 at a selected energy. The three coefficients give the intercept, slope, and curvature of the relative efficiency curve.

All of this information is used to determine the relative disintegration rates of the isotopes contributing to the selected peak intensities. The equations are nonlinear in several unknowns and thus require iterative, nonlinear least squares to solve. We have developed software to monitor and control the iterative process to insure that the mass thickness values for the absorber and sample stay within reasonable ranges. Initial estimates of the relative disintegration rates for the isotopes involved are made from a selected peak area for each isotope. The iterative process begins with the values for the isotope disintegration rates and the absorber thickness free. After four iterations the sample thickness is also freed. After seven iterations the absorber thickness is fixed, unless convergence is reached first. Once convergence is reached in the sample thickness, the efficiency slope is freed. When convergence is reached in the efficiency slope, the efficiency curvature is freed for two iterations. If convergence is not reached in twenty iterations for any of the thickness or efficiency curve values, the process stops and reports that convergence was not reached. When convergence is reached in the thickness and efficiency curve values, the relative amounts of the isotopes involved are calculated from the relative disintegration rates determined from the iterative process and the half-lives given by the user.

The equations are weighted according to the statistical uncertainties of the peak intensities. There are other sources of error that we have not included yet in our analysis, so the results given in this paper do not give calculated uncertainties. The results shown in Table II indicate the precision and accuracy that can be obtained.

3.0 MEASUREMENT RESULTS

We have tested this general approach to isotopic analysis on plutonium gamma rays above 200 keV from samples of high burnup plutonium. Such analyses would be required where the sample containers are thick enough to significantly reduce the intensities of gamma- and x-ray emissions below 200 keV. The spectra were taken with a coaxial, high-purity germanium detector with a detector efficiency of 25% at 1332-keV. We used the following gamma rays from seven different energy groupings to determine the relative abundances of the plutonium isotopes except Pu-242, which was declared for each analysis.

Table 1. Gamma-ray energies used to determine relative plutonium isotopic abundances

Energy (keV)	**Isotope**
203.6	Pu-239
208.0	U-237 - Pu-241, Am-241
332.3	U-237 – Pu-241, Am-241
345.0	Pu-239
375.0	Pu-239
376.6	Am-241
413.7	Pu-239
642.5	Pu-240
646.0	Pu-239
662.4	Am-241
721.9	Am-241
766.4	Pu-238
769.4	Pu-239

The detector cryostat had an up-looking configuration and the samples were placed on top of the detector with an intervening lead collimator, i.e. the measurements were made through the bottom of the sample container. Results from two sets of measurements of a 50-gram plutonium sample are given in Tables II and III.

Table II. Measurement results from four 1,000-second measurements of a 50-gram plutonium sample with a 1.3-cm thick steel bottom. An additional absorber of 6.0-mm of steel was used.

Declared	**Pu-238**	**Pu-239**	**Pu-240**	**Pu-241**	**Pu-242**
	1.721	58.096	24.768	9.771	5.645
Measured	1.72	60.68	26.65	10.06	
	1.73	60.53	26.73	10.04	
	1.76	60.76	26.42	10.08	
	1.75	60.78	26.42	10.08	
Average	1.74	60.69	26.55	10.06	
% Std. Dev.	1.05	0.19	0.60	0.19	
% Diff.	1.01	4.46	7.21	3.01	

Results shown in Table III are of the same sample with additional absorbers of cadmium, lead, and steel used in the measurements.

Table III. Measurement results from five measurements of the same sample used for the measurements shown in Table II. The measurement times and absorbers are shown for each measurement.

Declared	**Pu-238**	**Pu-239**	**Pu-240**	**Pu-241**	**Pu-242**
	1.721	58.096	24.768	9.771	5.645
Measured					
1,000 s 1.25 mm Cd 0.5 mm Pb	1.69	55.49	27.08	9.37	
1,000 s 1.25 mm Cd 0.5 mm Pb	1.78	56.89	26.07	9.61	
6,600 s 1.25 mm Cd 0.5 mm Pb	1.76	58.30	24.36	9.93	
1,000 s 1.25 mm Cd 0.5 mm Pb 4.0 mm Steel	1.77	56.43	26.71	9.44	
1,000 s 1.25 mm Cd 0.5 mm Pb 6.0 mm Steel	1.93	61.3	20.9	10.2	

4.0 SUMMARY

We utilize a unique and accurate methodology for delineating the relative detection efficiency that enables us to relate the intensities of widely separated gamma-ray peaks to determine relative abundances of specified isotopes. We have generalized the application of this methodology in software, so that the user can easily modify and adapt it to their analysis needs. The user may specify peak intensities determined by their own means, or they may specify how to fit peaks in a given energy region as described above. The user also specifies which peaks to use in an isotopic analysis along with their branching intensities. Finally, the user specifies the absorbers, and their thickness and compositions. The analysis software is very transparent with it using a set of equations that represent the physical processes that determine the measured gamma-ray intensities and physical parameters that are specified by the user.

Our testing has demonstrated that the methodology may be used to determine relative plutonium abundances from gamma rays above 200 keV. Our previous experience indicates that it can be used to determine uranium enrichment from gamma rays above 100 keV, and relative plutonium isotopic abundances in samples with enhanced amounts of Pu-238. Our software allows the user to adapt this methodology quickly and easily to almost any measurement configuration or isotopic analysis.

5.0 ACKNOWLEDGEMENTS

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